

Trees in urban parks and forests reduce O₃, but not NO₂ concentrations in Baltimore, MD, USA.

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Abstract

Trees and other vegetation absorb and capture air pollutants, leading to the common perception that they, and trees in particular, can improve air quality in cities and provide an important ecosystem service for urban inhabitants. Yet, there has been a lack of empirical evidence showing this at the local scale with different plant configurations and climatic regions. We studied the impact of urban park and forest vegetation on the levels of nitrogen dioxide (NO_2) and ground-level ozone (O_3) while controlling for temperature during early summer (May) using passive samplers in Baltimore, USA. Concentrations of O_3 were significantly lower in tree-covered habitats than in adjacent open habitats, but concentrations of NO_2 did not differ significantly between tree-covered and open habitats. Higher temperatures resulted in higher pollutant concentrations and NO_2 and O_3 concentration were negatively correlated with each other. Our results suggest that the role of trees in reducing NO_2 concentrations in urban parks and forests in the Mid-Atlantic USA is minor, but that the presence of tree-cover can result in lower O_3 levels compared to similar open areas. Our results further suggest that actions aiming at local air pollution mitigation should consider local variability in vegetation, climate, micro-climate, and traffic conditions.

Keywords: urban trees, air pollution, ecosystem services, urban vegetation, nitrogen dioxide, ozone

1. Introduction

Air pollution is amongst the most recognized environmental problems around the world.

Although concentrations of some air pollutants, such as nitrogen dioxide (NO_2) and ground-level ozone (O_3), have generally decreased within the past decades, their levels continue to exceed limits known to affect human health in many urban areas (Duncan et al., 2016; EEA, 2016; Pfister et al., 2014). The principal sources of NO_2 are energy production, industry and road traffic (EEA, 2016), but in cities nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$) are mostly emitted from traffic-related combustion as NO , which is quickly oxidized by O_3 to NO_2 - or directly as NO_2 (Anttila et al., 2011). The latter has been a growing trend worldwide, including USA, due to increasing proportion of modern diesel engines (Anenberg et al., 2017). Elevated NO_2 concentrations can cause an increase in respiratory symptoms and infections in asthmatic individuals and children (Kampa & Castanas, 2008) and result in increased prevalence of atopic sensitizations, allergic symptoms, and diseases (Krämer et al., 2000). Ground-level O_3 is formed in reactions where volatile organic compounds (VOCs) interact with NO_x in the presence of sunlight (Calfapietra et al., 2013; Loreto & Schnitzler, 2010). Ozone concentrations are thus dependent on both emissions of NO_x and VOCs. VOCs include anthropogenic sources such as traffic and manufacturing as well as natural compounds (biogenic VOCs) such as isoprene which are emitted by trees. In regions of high VOCs, ozone concentrations are highly sensitive to NO_x levels and vice versa. Urban and suburban populations are exposed to higher than ambient concentrations of ground-level O_3 especially during hot summer periods (Churkina et al., 2017) and in case of acute exposure inhabitants may suffer from reduced lung function and lung diseases (Uysal & Schapira, 2003).

A logical way to improve air quality is the reduction of emissions of air pollutants (Duncan et al., 2016; EEA, 2016), but it has been suggested that urban vegetation, especially trees, which can absorb and capture air pollutants with their large leaf area, could be also used to clean polluted urban air (Beckett et al., 2000; Nowak et al., 2006). For example, gases such as NO₂ (Chaparro-Suarez et al., 2011; Hu et al., 2016; Rondón & Granat, 1994; Takahashi et al., 2005) and O₃ (Hu et al., 2016; Manes et al., 2012; Wang et al., 2012) are absorbed from air through the stomata into the leaf interior of a plant. Such uptake of air pollutants by urban plants is often considered to result in effective ambient air quality improvement in city-scale and consequently to provide an important ecosystem service (e.g. Jim & Chen, 2008; Manes et al., 2012; Nowak et al., 2008). This has been emphasized especially in the interpretation of model studies (e.g. Baumgardner et al., 2012; Hirabayashi et al., 2012; Morani et al., 2011; Nowak et al., 2013; Selmi et al., 2016).

Recently, the overall significance of this ecosystem service has been challenged by contradictory results from local-scale studies and related critical comments (Churkina et al., 2017; Gromke & Ruck, 2009; Harris & Manning, 2010; Pataki et al., 2011; Pataki et al., 2013; Vos et al., 2013). Although studies comparing locally measured pollutant concentrations e.g. in urban forests and in adjacent open areas have been scarce, an increasing number of such studies have been published (Brantley et al., 2014; Fantozzi et al., 2015; Harris & Manning, 2010; Setälä et al., 2013; Tong et al., 2015; Viippola et al., 2016; Yin et al., 2011; Yli-Pelkonen et al., 2017). For instance, Setälä et al. (2013) and Yli-Pelkonen et al. (2017) did not find differences in gaseous pollutant concentrations between tree-covered and open near-road environments in hemi-boreal

climatic conditions, while Viippola et al. (2016) observed elevated gaseous PAH concentrations in road-side forests and parks compared to adjacent treeless areas during summer in Finland.

Some recent studies have used a city-wide measurement network of passive or active air collectors and combined this data with environmental variables such as land use, vegetation coverage and traffic. For instance, Rao et al. (2014) used a land-use regression model combined with NO₂ measurements at 144 sites in Portland, USA, and estimated a significant modelled NO₂ reduction due to tree canopy across the city. Irga et al. (2015) used portable active instruments for monthly air samples at eleven sites in Sydney, Australia, but found no observable trends in NO₂ concentrations between the sites with a range of different traffic and greenspace densities. Caballero et al. (2012) used passive samplers to study spatial and temporal variations of NO₂ levels at 79 sites in the city of Elche in Spain and observed that the spatial distribution of NO₂ depended mainly on the urban structure and traffic configuration, but the role of tree-cover on pollution levels could not be assessed with the study setup. García-Gómez et al. (2016) applied passive samplers and active monitors in studying O₃ and NO₂ levels in three peri-urban forests and one rural forest dominated by *Quercus ilex* and nearby open areas in Spain and found lower O₃ and NO₂ concentrations under tree canopies in the rural site and in one peri-urban site for O₃ and in two peri-urban sites for NO₂. Furthermore, Yin et al. (2011) studied six urban parks in Shanghai, China, and observed lower NO₂ concentrations inside parks with tree-cover as compared to a single reference site without tree-cover. Thus, the literature shows that different climatic conditions, plant configurations, degree of urbanization and the scale of a study area yield variable measurement results regarding the potential of urban vegetation to reduce the levels of gaseous air pollutants. This makes the interpretation of the modelling results even more

challenging and indicates the need for more empirical field-study data on the topic, that can also be used in model improvement.

Local temperature differences within a city and between tree-covered and open areas may also have an impact on air pollutant levels, as open areas receive more radiation that could influence photolysis reactions or heat the air locally, thereby affecting chemical reaction rates (Jacob, 1999). Temperature differences within a city can be related to land-use changes caused by urbanization. On an urban-rural scale, this difference is known as the Urban Heat Island (UHI) effect, but variability exists within the urban area as well. As with air quality, this variability can be linked to heterogeneity in urban form and has been linked with the presence or lack of vegetation and impervious surfaces (Scott et al., 2017). Thus, controlling for local variations in temperature is an important consideration.

The objective of our study was to explore the influence of urban tree-cover on the concentrations of gaseous air pollutants NO_2 and O_3 under early summertime conditions in Baltimore, MD, USA and thus provide much needed empirical evidence on the ability of urban vegetation to improve air quality. Based on previous findings from the above-mentioned studies, where the sampling sites were not in the close proximity to busy roads, we hypothesized that (1) air quality regarding the studied gaseous air pollutants in tree-covered urban areas is improved compared to adjacent open areas, and 2) the air quality improvement relates to temperature, amount of canopy cover and traffic volume.

2. Methods

2.1. Sampling

We measured the concentrations of NO₂ and ground-level O₃ using dry deposition passive collectors developed by the Swedish Environmental Research Institute IVL and temperature using 50 Maxim Integrated Products, Inc., “iButton” Model DS1923 Hygrochron thermometer/hygrometers. We installed the air collectors and thermometers either under tree canopies in tree-covered areas or in adjacent open areas in Baltimore, MD (39°17'57"N, 76°36'34"W), eastern USA (Fig. 1). NO₂ collectors and their analysis were provided by Metropolilab, Helsinki, Finland, and O₃ collectors and their analysis by IVL. The sampling of NO₂ and O₃ is based on molecular diffusion. The gas is adsorbed to a filter paper inside the collector and the amount of gas is analyzed by extracting it from the filter to distilled water, after which the amount of gas is determined with a spectrophotometer (Ayers et al., 1998). The NO₂ sampling method has been used successfully in numerous studies with results corroborated by active air monitoring instruments (Ayers et al., 1998; Ferm & Rodhe, 1997; Klingberg et al., 2017; Krupa & Legge, 2000; Caballero et al., 2012; HSY, 2014). The O₃ collectors have proved to be reliable and accurate according to our own tests alongside municipal active O₃ measuring instruments. iButton thermometers have an accuracy of 0.5 degrees C. The iButtons are shielded by a custom radiation shield that is naturally aspirated and made of White98 F-23, a commercial material manufactured by White Optics that is highly reflective for visible light and most often used in industrial lighting applications.

2.2. Sampling sites and dates

We established twenty-five sampling sites in urban parks and forests in Baltimore (Fig. 1). Four of the sites were situated in park-like block courtyards. At each site we installed air collectors and thermometers at the sampling points in two habitats: in a tree-covered area and in an adjacent open area. The tree-covered areas were as largely tree-covered around the sampling point as possible, but often included some non-canopy-covered area within a 50 m radius from the sampling point. Similarly, the open areas were as widely open around the sampling point as possible, but often included some amount of trees within a 50 m radius from the sampling point, usually situated close to the perimeter of this circle. Within each site, we situated the sampling points (open and tree-covered) at the approximately same distance from the nearest major road, but not in close proximity to heavily trafficked roads, as the idea was to study ambient urban air pollutant levels in green areas and not pollution coming from a single source. Some sites were situated within the urban street grid with low traffic streets. At different sites, depending on the availability of suitable mounting structures and the location of habitats, the distance between the sampling point pairs (open and tree-covered) and the nearest major road varied; ranging between 25 and 543 m (mean 136 m). The distance between the two sampling points (open or tree-covered) within each site ranged between 23 and 187 m (mean = 79.4 m). The tree-covered habitats consisted of mainly broadleaved, mature trees. The open habitats were mainly lawns or grasslands. The soil surface at these open habitats was either completely pervious or partly impervious with some asphalt surfaces.

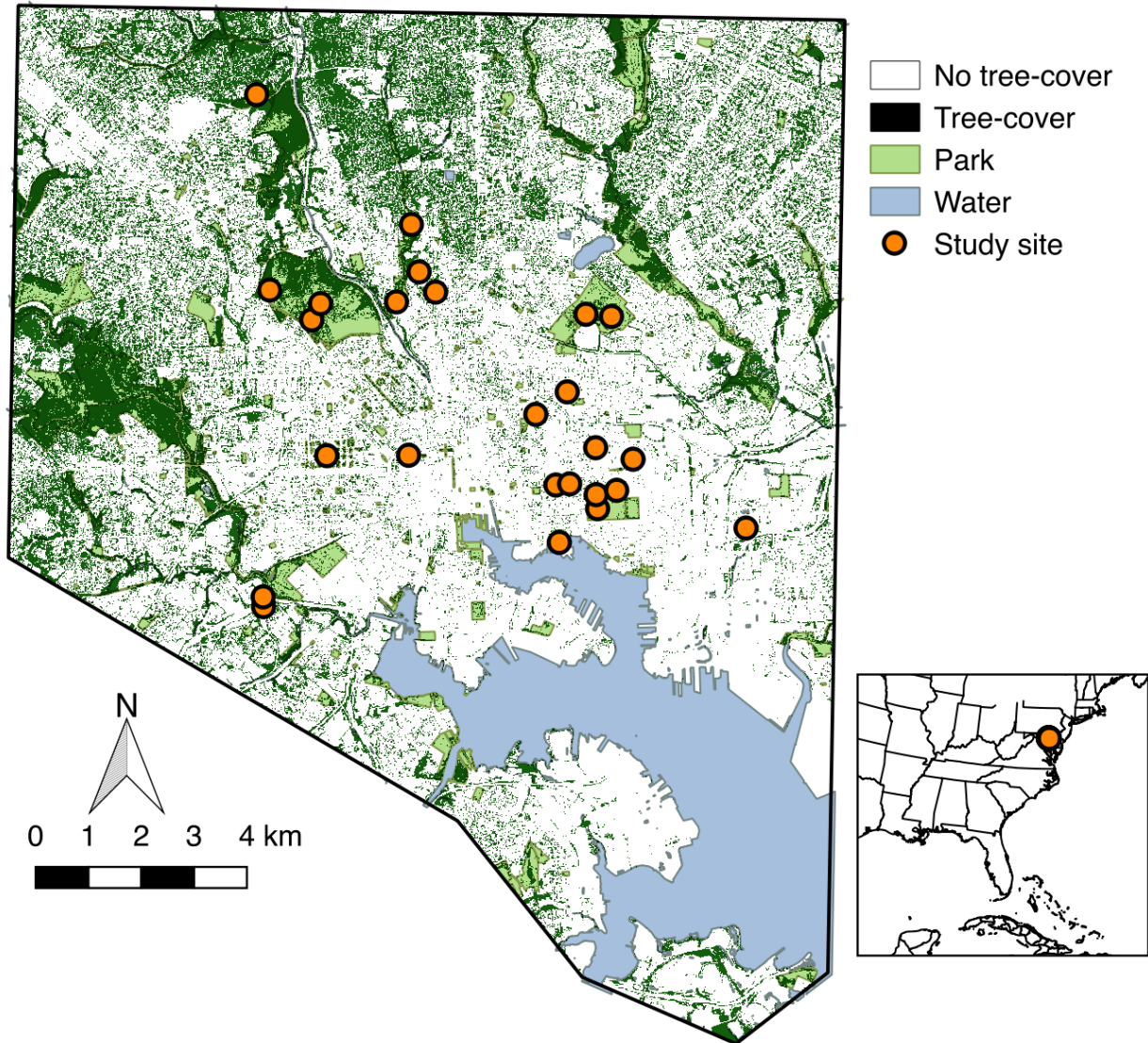


Figure 1. Locations of the twenty-five study sites (marked as dots) in Baltimore, MD, USA. The dot in a small map on the right depicts the location of greater Baltimore in USA. At each of the 25 sites, air quality and temperature was measured in both tree-covered and open habitats.

We mounted the air samplers under rain shields and the thermometers inside radiation shields and attached them to lamp posts or similar structures in the open areas and to tree trunks (directly under the canopy) in the tree-covered areas. We mounted the air samplers and thermometers 3-4

m above ground to prevent samplers being too close to ground surface where O₃ gets depleted (Mills et al., 2010), yet representing the height in which humans are exposed to these pollutants. Measurements were carried out during early summer, from 5 May to 25 May, 2016 (20 days), when plant leaves were practically fully developed.

We determined the percentage of canopy cover at each site from satellite images (using Google Earth Pro, version 7.1.2.2041) by measuring the area covered by tree canopy within 50 m radii from the sampling point in both habitats. The percentage of canopy cover ranged between 25 and 100 % (mean = 63.9 %) in tree-covered habitats and between 0 and 73 % (mean = 23.8 %) in open habitats. Mean daily temperatures ranged between 14.5 and 16.3 °C (mean = 15.5 °C) in tree-covered habitats and between 15.4 and 17.5 °C (mean = 16.3 °C) in open habitats, being significantly lower in the tree-covered habitats than in the open habitats (5.2%, $p < 0.001$, $n = 23$ due to two lost thermometers). We determined traffic volume at each site by calculating the cumulative traffic volume from the largest streets within 200 m radii from the sampling point in the tree-covered area using traffic flow data (annual average daily traffic), obtained from Baltimore Metropolitan Council (2016) and Maryland Department of Transportation (2016). Traffic volume (motor vehicles day⁻¹) ranged between 0 and 179,066 (mean = 36,138). We did not estimate traffic volume separately for open habitat sampling points as the open and tree-covered habitat circles with 200 m radii would overlap to such extent that traffic volume would practically be the same.

The sampling sites situated in remnant forest patches, parks and park-like block courtyards in residential areas were dominated by broad-leaf deciduous trees, including tree species (*Fraxinus*

spp., *Ulmus americana*, *Fagus grandifolia*, *Prunus serotina*, *Robinia pseudoacacia* and *Ailanthus altissima*) typical to Baltimore City. The sampling sites situated in the outskirts of the city additionally included dominant forest tree species (broad-leaf deciduous) (*Quercus montana*, *Liriodendron tulipifera*, *Acer negundo*, *Fraxinus pennsylvanica*, *Platanus occidentalis* and *Acer saccharinum*) typical to forests outside of the city of Baltimore. A wind rose showing the prevailing wind direction and speed during the measuring period is shown in Fig. 2. The monthly average temperature in Baltimore in May 2016 was 16.0 °C, representing slightly lower temperatures than usually in May in Baltimore.

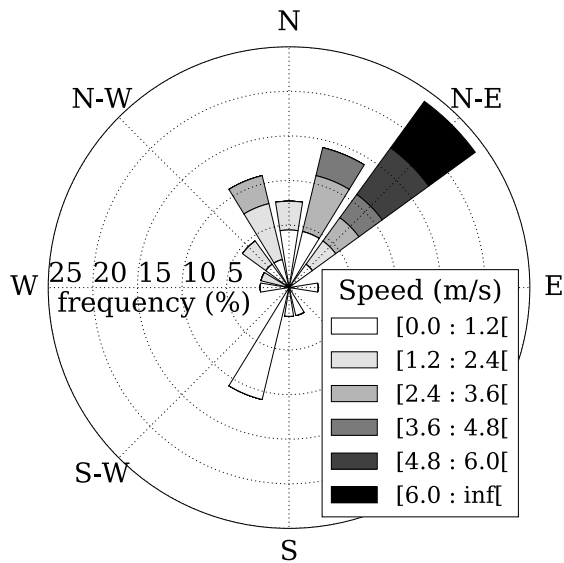


Figure 2. Wind direction and speed during 5 May – 25 May, 2016 in Baltimore (Johns Hopkins Homewood Campus measuring station).

2.3. Data analysis

We tested changes in NO₂ and O₃ concentrations using linear models, with NO₂ and O₃ modelled following a normal distribution. Two types of analyses were performed. First, we evaluated the concentrations of NO₂ and O₃ across the whole dataset using the following model structure;

NO₂ ~ habitat type + O₃ + mean daily temperature, and

O₃ ~ habitat type + NO₂ + mean daily temperature.

Here habitat type represents a factor with two levels; tree-covered and open areas. Second, we evaluated the concentrations of NO₂ and O₃ per habitat type. This resulted in four analyses;

NO_{2 tree} ~ traffic volume + canopy cover_{tree} + O_{3 tree} + mean daily temperature_{tree}

NO_{2 open} ~ traffic volume + canopy cover_{open} + O_{3 open} + mean daily temperature_{open}

O_{3 tree} ~ traffic volume + canopy cover_{tree} + NO_{2 tree} + mean daily temperature_{tree}

O_{3 open} ~ traffic volume + canopy cover_{open} + NO_{2 open} + mean daily temperature_{open}

The subscripts above relate to the measurements in that particular habitat type. Traffic volume was square-root transformed into a normal distributed variable, due to a few very high values.

Model selection was performed in all six models by removing predictors, one at a time, if their p-values were greater than 0.1. All data analyses were performed using R statistical software, version 3.4 (R Core Team, 2017).

3. Results

While the mean NO₂ concentration appears lower in tree-covered areas than in open areas (Fig. 3), the difference is not statistically significant (Table 1). However, O₃ concentrations were significantly lower in tree-covered areas compared to open areas (Table 1, Fig. 3). Across the whole dataset, both NO₂ and O₃ levels were significantly positively related with mean daily temperature, and NO₂ and O₃ levels were significantly negatively correlated with each other (Table 1). Traffic volume was retained in all four models dealing with NO₂ and O₃ per habitat type. NO₂ in both the tree-covered and open areas increased significantly with traffic volume, while O₃ in both the tree-covered and open areas decreased significantly with traffic volume (Fig. 4, Table 1). Additionally, NO₂ concentrations in the tree-covered areas increased significantly with temperature and NO₂ concentrations in the open areas decreased significantly with the percentage of canopy cover (Fig. 4, Table 1).

Table 1. Linear model results (see Figs. 3 and 4), testing the effects of various variables on NO₂ and O₃ levels across the whole dataset and per habitat type. The subscripts (_{tree} or _{open}) relate to the measurements in that particular habitat type. Coefficients with standard errors (in brackets) and p-values (below standard errors) are presented. The intercept included open habitat type in the first two models (whole dataset models).

Variable		Intercept	Habitat type	NO ₂ / O ₃	Mean daily temperature	Canopy cover	Traffic volume
NO ₂	Coefficient	-33.787		-0.315	4.490		
	SE	(11.874)		(0.123)	(0.825)		
	<i>p</i>	0.007		0.014	<0.001		
O ₃	Coefficient	24.363	-3.997	-0.337	2.382		
	SE	(18.140)	(1.437)	(0.156)	(1.212)		
	<i>p</i>	0.186	0.008	0.036	0.056		

NO ₂ tree	Coefficient	-67.769	5.501	0.016
	SE	(16.614)	(1.080)	(0.004)
	<i>p</i>	<0.001	<0.001	0.001
NO ₂ open	Coefficient	25.558	-0.002	0.030
	SE	(1.613)	(<0.001)	(0.004)
	<i>p</i>	<0.001	<0.001	<0.001
O ₃ tree	Coefficient	53.364		-0.019
	SE	(1.194)		(0.006)
	<i>p</i>	<0.001		0.006
O ₃ open	Coefficient	57.579		-0.012
	SE	(1.134)		(0.006)
	<i>p</i>	<0.001		0.051

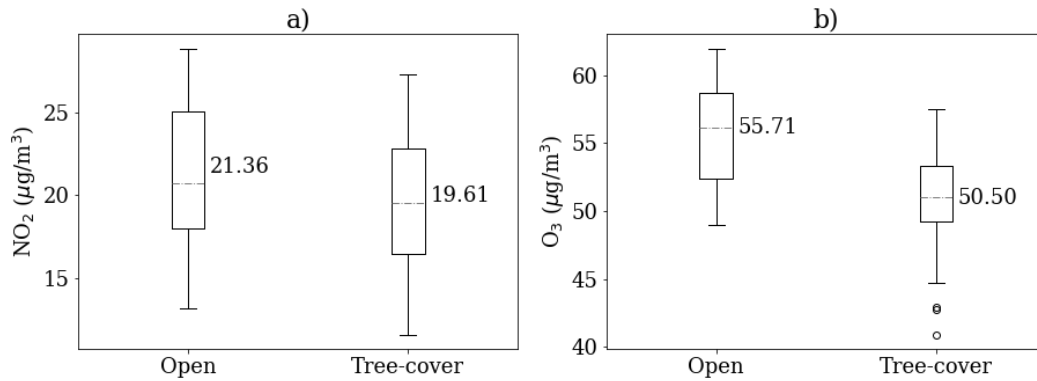


Figure 3. Concentrations of (a) NO₂, and (b) O₃ in open and tree-covered areas ($n = 25$). The dashed grey line indicates the mean (labelled), the box indicates the first through third quartiles, and whiskers delineate the wide interquartile range, 1.5 times the first through third quartiles. Data points falling outside this range are shown as dots.

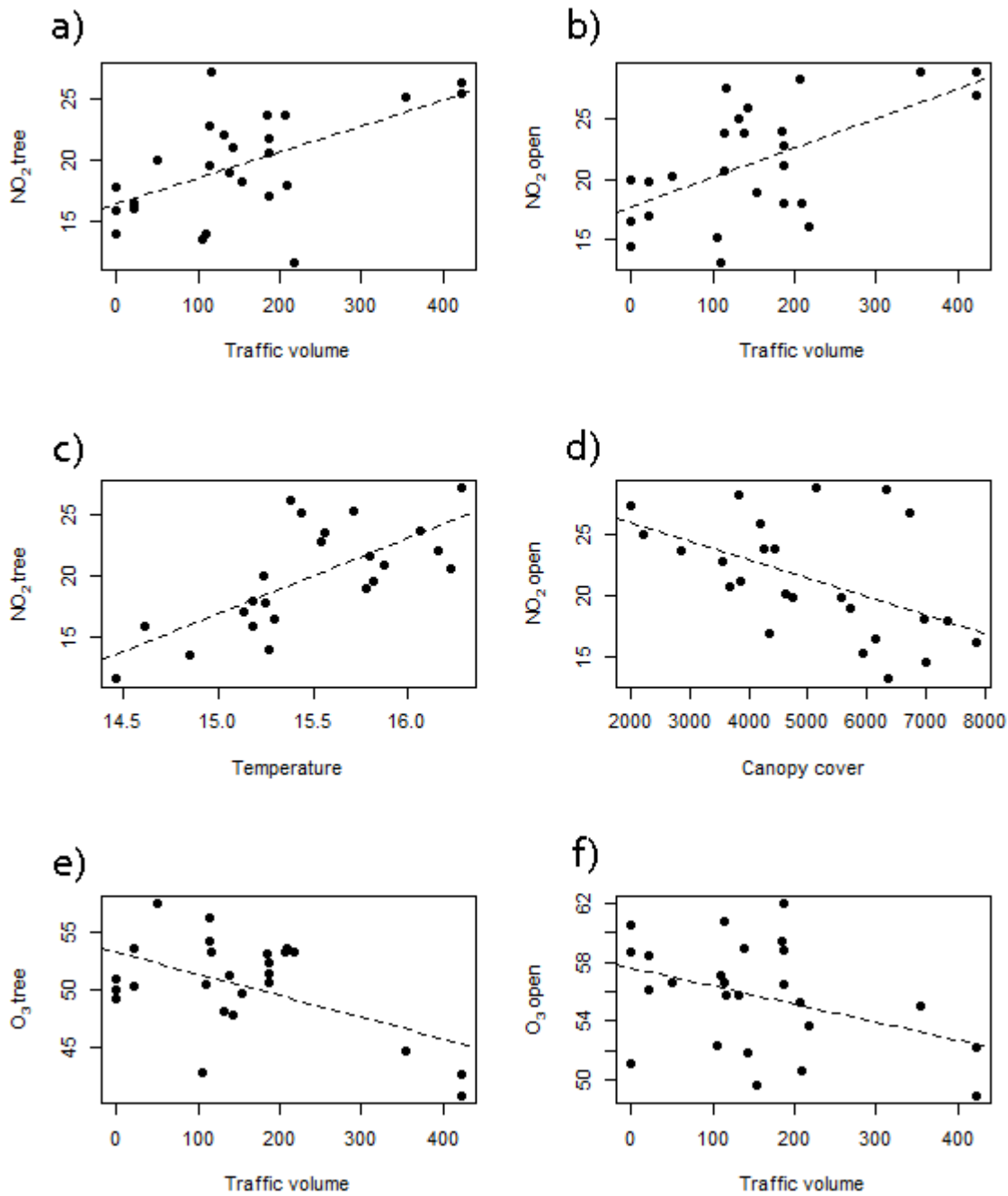


Figure 4. Relations between (a) NO_2 concentrations in tree-covered areas and traffic volume (square-root transformed number of motor vehicles day^{-1} , annual average of daily traffic, presented within 200 m radius from the sampling point in the tree-covered area); (b) NO_2 concentrations in open areas and traffic volume; (c) NO_2 concentrations in tree-covered areas

and mean daily temperatures (C°) in tree-covered areas; (d) NO_2 concentrations in open areas and canopy cover in open areas (m^2 , within the 50 m radius from the sampling point); (e) O_3 concentrations in tree covered-areas and traffic volume, and (f) O_3 concentrations in open areas and traffic volume. See Table 1 for statistical results.

4. Discussion

Our study, performed in early summertime (May) in urban forest/park environments in the Mid-Atlantic USA, suggests that the concentrations of NO_2 are not significantly reduced in tree-covered habitats compared to adjacent open habitats, but that O_3 concentrations are. That NO_2 concentrations did not differ between tree-covered and open habitats is in contrast to our hypothesis and those earlier empirical studies (García-Gómez et al., 2016; Grundström & Pleijel, 2014; Fantozzi et al., 2015; Klingberg et al., 2017; Rao et al., 2014; Yin et al., 2011), where some NO_2 reduction by tree-canopy was detected. However, our finding that NO_2 concentrations in the open habitats decreased with the increasing percentage of canopy cover (within a 50 m radius from the sampling point in the open habitat) may indicate some absorption of NO_2 by the trees. On the contrary, our result that NO_2 levels were not reduced in tree-covered habitats corroborates those empirical findings, where no clear reduction of NO_2 levels by urban tree cover was observed either on city-level (Irga et al., 2015) or on local level in near-road environments (Setälä et al., 2013; Yli-Pelkonen et al., 2017), or where even higher concentrations of NO_2 were reported inside the urban tree canopies than outside (Harris & Manning, 2010). Our findings showing air quality improvement by urban tree-cover in terms of O_3 support our hypothesis and similar earlier empirical findings (García-Gómez et al., 2016;

Harris & Manning, 2010) and a number of city-scale modeling studies (e.g. Baumgardner et al., 2012; Selmi et al., 2016), but are partly in contrast to local-level findings of e.g Grundström & Pleijel (2014), Fantozzi et al. (2015) and Yli-Pelkonen et al. (2017). Although data on O₃ uptake rates by all the tree species in our study area is not available, broad-leaf deciduous trees are generally estimated to be relatively efficient in O₃ removal (Manes et al., 2012).

For instance, Harris & Manning (2010) used passive samplers and temperature loggers in Springfield, Massachusetts, USA, and found higher NO₂ and lower O₃ levels inside urban tree canopies than directly outside (30-45 cm) the canopies, at locations with varying distance from a major highway. They suggested that this is due to NO_x/O₃ chemistry related to gas interactions between soil and the air, as described by Fowler (2002). However, they did not find differences in temperature between inner and outer canopy locations and suggested that temperature had little or no effect on the pollutant levels. In contrast, our open sites were much farther from tree canopies and shade, and grow hotter during the day. Fantozzi et al. (2015) studied one site in Siena, central Italy, and found lower NO₂ concentrations in a measurement transect under the canopies of *Quercus ilex* L., extending 1-10 m from a busy road, compared to a nearby open-field transect. They observed reduced O₃ concentrations inside the canopy transect only during post-summer rainfalls. Yli-Pelkonen et al. (2017) measured NO₂ and O₃ concentrations on average 25 m from busy roads at 10 sites in Helsinki, Finland, where NO₂ concentrations were on average similar to and O₃ concentrations clearly lower than in our study in Baltimore, but did not find differences between tree-covered and adjacent open road-side habitats. The variable results from different parts of the world indicate that the impact of vegetation on the concentration of gaseous air pollutants such as NO₂ and O₃ may largely depend on the type and

structure of local vegetation, climatic conditions, proximity to traffic pollution sources and regional and local ambient pollutant levels.

It is well established that formation of O₃ needs photolysis and is positively related to temperature (Cardelino & Chameides, 1990; Churkina et al., 2017; Finlayson-Pitts & Pitts, 1997; Paoletti et al., 2014), which was also detected in our study. As it is also likely that increased shade and the observed lower mean daily temperatures within the tree-canopies also reduce solar radiation under the canopies (Shashua-Bar & Hoffman, 2004; Renaud et al., 2011; Lehmann et al., 2014), this may result in decreased concentrations of O₃ in tree-covered habitats compared to open habitats (Cardelino & Chameides, 1990). Thus, the observed lower O₃ concentrations in the tree-covered habitats may not result solely from the O₃ uptake by tree canopy, but also from the reduced solar radiation and temperatures. That NO₂ levels in our study decreased with temperature across the whole dataset and in the tree-covered habitats may also indicate a slight NO₂ uptake by the tree canopies, which were cooler than open areas. However, as NO₂ levels did not differ significantly between tree-covered and open habitats, the reason for the weak relationship between NO₂ concentration and temperature remains unsolved.

Traffic volume's positive relation to NO₂ levels and negative relation to O₃ levels, as well as the negative relation between NO₂ and O₃ levels, indicate that NO₂ in the study area originates mainly from road traffic (see e.g. Clements et al., 2009; Setälä et al., 2013; Yli-Pelkonen et al., 2017) as higher traffic volume produces more NO_x. While at low concentrations, NO_x is a precursor to ozone formation, at higher concentrations (all else being equal), NO_x catalyzes ozone destruction, which results in O₃ depletion (e.g. Rodes & Holland, 1981). In all, the

interplay between traffic density, NO₂ and O₃ depends on an array of factors in urbanized settings, even at sites situated far from heavily-trafficked roads.

The prevailing wind direction (long-term average) in May in Baltimore is from the south, but during the measuring period north-eastern winds dominated (Fig. 2). However, as our sampling sites were relatively far away from heavily-trafficked roads or neutrally situated within the street network, we did not place the sampling sites according to the prevailing wind direction.

Furthermore, our relatively long sampling period is bound to diminish the impacts of short-term wind direction changes and thus the wind direction is unlikely to cause systematic bias in our measurement campaign.

As has been noted in earlier studies, it is possible that reduced air flow within tree-covered areas in near-road environments (Belcher et al., 2012; Gromke & Ruck, 2009; Renaud et al., 2011; Wuyts et al., 2008) can increase pollutant levels within the canopy and thus have negative impacts on local air quality (Setälä et al., 2013; Viippola et al., 2016; Vos et al., 2013), while in open areas the polluted air mass is mixed by wind and diluted more rapidly. However, as in the current study in Baltimore the sampling sites were not in close proximity to heavily-trafficked roads, such "trapping effect" of highly polluted air mass under the canopies is unlikely and the actual uptake of the studied gaseous pollutants by tree canopies should be observable.

Our results suggest that one should not take for granted the notion that urban trees provide overall air quality benefits. For instance, NO₂ concentrations are not necessarily decreased in the tree-covered areas via absorption of NO₂ by trees. However, tree-cover seems to provide air

quality improvement regarding O_3 , at least in the early summer conditions in Baltimore. This may not be the case during the heat wave periods in mid-summer, when elevated emissions of biogenic volatile organic compounds could, in combination with NO_x emissions from traffic, contribute to increased O_3 formation in urban areas (Churkina et al., 2017; Manes et al., 2012). However, as tree species in the study area are not strong emitters of reactive biogenic volatile organic compounds, their ozone forming potential is minor (Benjamin & Winer, 1998).

5. Conclusions

Our results, obtained from the period when gas exchange between foliage and the atmosphere is active in Baltimore, Mid-Atlantic USA, suggest that trees in urban forests and parks can result in reduced O_3 levels, but that the impact of trees on NO_2 concentrations is negligible. That O_3 concentrations were reduced under tree canopies was in line with our hypothesis and likely results from absorption of O_3 by tree canopies, or from lowered temperatures and reduced solar radiation under tree canopies - or combination of these. That NO_2 concentrations did not differ between tree-covered and open habitats was in contrast to our hypothesis and suggests that urban park and forest vegetation do not necessarily provide significant and uniform air quality improvement regarding NO_2 , as it is often stated by city-scale model calculations.

Thus, urban inhabitants spending time in tree-covered areas in urban parks and forests may receive lower exposure to O_3 concentrations, at least when temperatures are moderate, but the same conclusion does not hold for NO_2 . We stress that trees and other vegetation provide many other ecosystem services for urban dwellers to enjoy, such as reduced temperatures under tree

canopies. Our results further suggest that actions aiming at local air pollution mitigation should consider local variability in vegetation, climate, micro-climate, and traffic conditions. We conclude that the key measure to reduce human exposure to NO₂ and ozone, should be reduction of emissions from traffic, and placing recreational areas far from pollutant sources.

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References

- Anenberg, S.C., Miller, J., Minjares, R., Du, L., Henze, D.K., Lacey, F., Malley, C.S., Emberson, L., Franco, V., Klimont, Z., Heyes, C., 2017. Impacts and mitigation of excess diesel-related NO_x emissions in 11 major vehicle markets. *Nature* 545, 467–471.
- Anttila, P., Tuovinen, J.-P., Niemi, J.V., 2011. Primary NO₂ emissions and their role in the development of NO₂ concentrations in a traffic environment. *Atmospheric Environment* 45, 986–992.
- Ayers, G.P., Keywood, M.D., Gillett, R., Manins, P.C., Malfroy, H., Bardsley, T., 1998. Validation of passive diffusion samplers for SO₂ and NO₂. *Atmospheric Environment* 32, 3587–3592.

Baltimore Metropolitan Council, 2016. Traffic Count Map. Retrieved 7 December, 2016 from - <http://www.baltometro.org/information-center/maps-and-data/interactive-mapping>

Baumgardner, D., Varela, S., Escobedo, F.J., Chacalo, A., Ochoa, C., 2012) The role of peri-urban forest on air quality improvement in the Mexico City megalopolis. *Environmental Pollution* 163, 174–183.

Beckett, K.P., Freer-Smith, P.H., Taylor, G., 2000. The capture of particulate pollution by trees at five contrasting urban sites. *Arboricultural Journal* 24, 209–230.

Belcher, S.E., Harman, I.N., Finnigan, J.J., 2012. The wind in the willows: flows in forest canopies in complex terrain. *Annual Review of Fluid Mechanics* 44, 479–504.

Benjamin, M.T., Winer, A.M., 1998. Estimating the ozone-forming potential of urban trees and shrubs. *Atmospheric Environment* 32, 53–68.

Brantley, H.L., Hagler, G.S.W., Deshmukh, P.J., Baldauf, R.W., 2014. Field assessment of the effects of roadside vegetation on near-road black carbon and particulate matter. *Science of the Total Environment* 468-469, 120–129.

Caballero, S., Esclapez, R., Galindo, N., Mantilla, E., Crespo, J., 2012. Use of a passive sampling network for the determination of urban NO₂ spatiotemporal variations. *Atmospheric Environment* 63, 148–155.

Calfapietra, C., Fares, S., Manes, F., Morani, A., Sgrigna, G., Loreto, F., 2013. Role of Biogenic Volatile Compounds (BVOC) emitted by urban trees on ozone concentration in cities: A review. *Environmental Pollution* 183, 71–80.

- Cardelino, C.A., Chameides, W.L., 1990. Natural hydrocarbons, urbanization, and urban zone. *Journal of Geophysical Research* 95, 13971–13979.
- Chaparro-Suarez, I.G., Meixner, F.X., Kesselmeier, J., 2011. Nitrogen dioxide (NO₂) uptake by vegetation controlled by atmospheric concentrations and plant stomatal aperture. *Atmospheric Environment* 45, 5742–5750.
- Churkina, G., Kuik, F., Bonn, B., Lauer, A., Grote, R., Tomiak, K., Butler, T.M., 2017. Effect of VOC emissions from vegetation on air quality in Berlin during a heatwave. *Environmental Science & Technology* 51, 6120–6130.
- Clements, A.L., Jia, Y., Denbleyker, A., McDonald-Buller, E., Fraser, M.P., Allen, D.T., Collins, D.R., Michel, E., Pudota, J., Sullivan, D., Zhu, Y., 2009. Air pollutant concentrations near three Texas roadways, Part II: Chemical characterization and transformation of pollutants. *Atmospheric Environment* 43, 4523–4534.
- Duncan, B.N., Lamsal, L.N., Thompson, A.M., Yoshida, Y., Lu, Z., Streets, D.G., Hurwitz, M.M., Pickering, K.E., 2016. A space-based, high-resolution view of notable changes in urban NO_x pollution around the world (2005-2014). *Journal of Geophysical Research* 121: 976–996.
- EEA, 2016. Air quality in Europe – 2016 report. European Environment Agency EEA Report No 28/2016. Retrieved 2 December, 2016 from - <http://www.eea.europa.eu/publications/air-quality-in-europe-2016>
- Fantozzi, F., Monaci, F., Blanusa, T., Bargagli, R., 2015. Spatio-temporal variations of ozone and nitrogen dioxide concentrations under urban trees and in a nearby open area. *Urban Climate*, 12, 119–127.

- Ferm, M., Rodhe, H., 1997. Measurements of air concentrations of SO₂, NO₂ and NH₃ at rural and remote sites in Asia. *Journal of Atmospheric Chemistry* 27, 17–29.
- Finlayson-Pitts, B.J., Pitts, J.N. Jr., 1997. Tropospheric air pollution: ozone, airborne toxics, polycyclic aromatic hydrocarbon, and particles. *Science* 276, 1045–1052.
- Fowler, D., 2002. Pollutant Deposition and Uptake by Vegetation, in: Bell J.N.B. & Treshow M. (Eds.), *Air Pollution and Plant Life*, John Wiley and Sons, Ltd., New York, pp. 43–67
- García-Gómez, H., Aguilera, L., Izquierda-Rojano, S., Valiño, F., Àvila, A., Elustondo, D., Santamaría, J.M., Alastuey, A., Calvete-Sogo, H., Gonzáles-Fernández, I., Alonso, R., 2016. Atmospheric pollutants in peri-urban forests of *Quercus ilex*: evidence of pollution abatement and threats for vegetation. *Environmental Science and Pollution Research* 23, 6400–6413.
- Gromke, C., Ruck, B., 2009. On the impact of trees on dispersion processes of traffic emissions in street canyons. *Boundary-Layer Meteorology* 131, 19–34.
- Grundström, M., Pleijel, H., 2014. Limited effect of urban tree vegetation on NO₂ and O₃ concentrations near a traffic route. *Environmental Pollution* 189, 73–76.
- Harris, T.B., Manning, W.J., 2010. Nitrogen dioxide and ozone levels in urban tree canopies. *Environmental Pollution* 158, 2384–2386.
- Hirabayashi, S., Kroll, C.N., Nowak, D.J., 2012. Development of a distributed air pollutant dry deposition modeling framework. *Environmental Pollution* 171, 9–17.
- Hu, Y., Zhao, P., Niu, J., Sun, Z., Zhu, L., Ni, G., 2016. Canopy stomatal uptake of NO_x, SO₂ and O₃ by mature urban plantations based on sap flow measurement. *Atmospheric Environment* 125, 165-177.

HSY (Helsinki Region Environmental Services Authority), 2014. Ilmanlaatu pääkaupunkiseudulla vuonna 2013 (Air Quality in the Helsinki Metropolitan Area in 2013). HSY publications 3/2014. [in Finnish with an abstract in English] Retrieved 5 September, 2016 from - https://www.hsy.fi/sites/Esitteet/EsitteetKatalogi/Julkaisusarja/3_2014_Ilmanlaatu_paakaupunkiseudulla_2013.pdf

Irga, P.J., Burchett, M.D., Torpy F.R., 2015. Does urban forestry have a quantitative effect on ambient air quality in an urban environment? *Atmospheric Environment* 120, 173–181.

Jacob, D., 1999. Introduction to atmospheric chemistry. Princeton University Press.

Jim, C.Y., Chen, W.Y., 2008. Assessing the ecosystem service of air pollutant removal by urban trees in Guangzhou (China). *Journal of Environmental Management* 88, 665–676.

Kampa, M., Castanas, E., 2008. Human health effects of air pollution. *Environmental Pollution* 151, 362–367.

Klingberg, J., Broberg, M., Strandberg, B., Thorsson, P., Pleijel, H., 2017. Influence of urban vegetation on air pollution and noise exposure – A case study in Gothenburg, Sweden. *Science of the Total Environment* 599-600, 1728–1739.

Krupa, S.V., Legge, A.H., 2000. Passive sampling of ambient, gaseous air pollutants: An assessment from an ecological perspective. *Environmental Pollution* 107, 31–45.

Krämer, U., Koch, T., Ranft, U., Ring, J., Behrendt, H., 2000. Traffic-related air pollution is associated with atopy in children living in urban areas. *Epidemiology* 11, 64–70.

Lehmann, I., Mathey, J., Rössler, S., Bräuer, A., Goldberg, V., 2014. Urban vegetation structure types as a methodological approach for identifying ecosystem services - Application to the analysis of micro-climatic effects. *Ecological Indicators* 42, 58–72.

Loreto, F., Schnitzler, J.-P., 2010. Abiotic stresses and induced BVOCs. *Trends in Plant Science* 15, 154–166.

Manes, F., Incerti, G., Salvatori, E., Vitale, M., Ricotta, C., Costanza, R., 2012. Urban ecosystem services: tree diversity and stability of tropospheric ozone removal. *Ecological Applications* 22, 349–360.

Maryland Department of Transportation, 2016. State Highway Administration, Traffic Monitoring Systems. Retrieved 7 December, 2016 from -
<http://sha.md.gov/Index.aspx?PageId=792>

Mills, G., Pleijel, H., Büker, P., Braun, S., Emberson, L., Harmens, H., Hayes, F., Simpson, D., Grünhage, L., Karlsson, P.-E., Danielsson, H., Bermejo, V., Gonzalez Fernandez, I. (Eds.), 2010. Chapter 3 of the LRTAP Convention Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads & Levels and Air Pollution Effects, Risks and Trends. Retrieved 5 September, 2016 from -
<http://www.rivm.nl/media/documenten/cce/manual/Ch3revisedsummer2010updatedJune2011.pdf>

Morani, A., Nowak, D.J., Hirabayashi, S., Calfapietra, C., 2011. How to select the best tree planting locations to enhance air pollution removal in the MillionTreesNYC initiative. *Environmental Pollution* 159, 1040–1047.

- Nowak, D.J., Crane, D.E., Stevens, J.C., 2006. Air pollution removal by urban trees and shrubs in the United States. *Urban Forestry & Urban Greening* 4, 115–123.
- Nowak, D.J., Crane, D.E., Stevens, J.C., Hoehn, R.E., Walton, J.T., Bond, J., 2008. A ground-based method of assessing urban forest structure and ecosystem services. *Arboriculture & Urban Forestry* 34, 347–358.
- Nowak, D.J., Hirabayashi, S., Bodine, A., Hoehn, R., 2013. Modeled PM_{2.5} removal by ten U.S. cities and associated health effects. *Environmental Pollution* 178, 395–402.
- Paoletti, E., De Marco, A., Beddows, D.C.S., Harrison, R.M., Manning, W.J., 2014. Ozone levels in European and USA cities are increasing more than at rural sites, while peak values are decreasing. *Environmental Pollution* 192, 295–299.
- Pataki, D.E., Alberti, M., Cadenasso, M.L., Felson, A.J., McDonnell, M.J., Pincetl, S., Pouyat, R.V., Setälä, H., Whitlow, T.H., 2013. City trees: Urban greening needs better data. *Nature* 502, 624.
- Pataki, D.E., Carreiro, M.M., Cherrier, J., Grulke, N.E., Jennings, V., Pincetl, S., Pouyat, R.V., Whitlow, T.H., Zipperer, W.C., 2011. Coupling biogeochemical cycles in urban environments: ecosystem services, green solutions, and misconceptions. *Frontiers in Ecology and the Environment* 9, 27–36.
- Pfister, G.G., Walters, S., Lamarque, J.-F., Fast, J., Barth, M.C., Wong, J., Done, J., Holland, G., Bruyère, C.L., 2014. Projections of future summertime ozone over the U.S. *Journal of Geophysical Research* 119, 5559–5582.

R Core Team, 2017. R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria. Retrieved 1 June, 2017 from - <https://www.R-project.org/>

Rao, M., George, L.A., Rosenstiel, T.N., Shandas, V., Dinno, A., 2014. Assessing the relationship among urban trees, nitrogen dioxide, and respiratory health. *Environmental Pollution* 194, 96–104.

Renaud, V., Innes, J.L., Dobbertin, M., Rebetez, M., 2011. Comparison between open-site and below-canopy climatic conditions in Switzerland for different types of forests over 10 years (1998-2007). *Theoretical and Applied Climatology* 105, 119–127.

Rodes, C.E., Holland, D.M., 1981. Variations of NO, NO₂ and O₃ concentrations downwind of a Los Angeles freeway. *Atmospheric Environment* 15, 243–250.

Rondón, A., Granat, L., 1994. Studies on the dry deposition of NO₂ to coniferous species at low NO₂ concentrations. *Tellus* 46B, 339–352.

Scott, A.A., Zaitchik, B., Waugh, D.W., O'Meara, K., 2017. Intraurban temperature variability in Baltimore. *Journal of Applied Meteorology and Climatology* 56, 159–171.

Selmi, W., Weber, C., Rivière, E., Blond, N., Mehdi, L., Nowak, D., 2016. Air pollution removal by trees in public green spaces in Strasbourg city, France. *Urban Forestry & Urban Greening* 17, 192–201.

Setälä, H., Viippola, V., Rantalainen, A.-L., Pennanen, A., Yli-Pelkonen, V., 2013. Does urban vegetation mitigate air pollution in northern conditions? *Environmental Pollution* 183, 104–112.

Shashua-Bar, L., Hoffman, M.E., 2004. Quantitative evaluation of passive cooling of the UCL microclimate in hot regions in summer, case study: urban streets and courtyards with trees.

Building and Environment 39, 1087–1099.

Takahashi, M., Higaki, A., Nohno, M., Kamada, M., Okamura, Y., Matsui, K., Kitani, S., Morikawa, H., 2005. Differential assimilation of nitrogen dioxide by 70 taxa of roadside trees at an urban pollution level. *Chemosphere* 61, 633–639.

Tong, Z., Whitlow, T. H., MacRae, P. F., Landers, A. J., Harada, Y., 2015. Quantifying the effect of vegetation on near-road air quality using brief campaigns. *Environmental Pollution*, 201, 141–149.

Uysal, N., Schapira, R.M., 2003. Effects of ozone on lung function and lung diseases. *Current Opinion in Pulmonary Medicine* 8, 144–150.

Viippola, V., Rantalainen, A.-L., Yli-Pelkonen, V., Tervo, P., Setälä, H., 2016. Gaseous polycyclic aromatic hydrocarbon concentrations are higher in urban forests than adjacent open areas during summer but not in winter - Exploratory study. *Environmental Pollution* 208, 233–240.

Vos, P.E.J., Maiheu, B., Vankerkom, J., Janssen, S., 2013. Improving local air quality in cities: To tree or not to tree? *Environmental Pollution* 183, 113–122.

Wang, H., Zhou, W., Wang, X., Gao, F., Zheng, H., Tong, L., Ouyang, Z., 2012. Ozone uptake by adult urban trees based on sap flow measurement. *Environmental Pollution* 162, 275–286.

Wuyts, K., Verheyen, K., De Schrijver, A., Cornelis, W. M., Gabriels, D., 2008. The impact of forest edge structure on longitudinal patterns of deposition, wind speed, and turbulence.

Atmospheric Environment 42, 8651–8660.

Yin, S., Shen, Z., Zhou, P., Zou, X., Che, S., Wang, W., 2011. Quantifying air pollution attenuation within urban parks: An experimental approach in Shanghai, China. Environmental

Pollution 159, 2155–2163.

Yli-Pelkonen, V., Setälä, H., Viippola, V., 2017. Urban forests near roads do not reduce gaseous air pollutant concentrations but have an impact on particles levels. Landscape and Urban

Planning 158, 39–47.